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This paper will give a general overview/problem definition and state-of-the-art of the natural processes of biodegradation and chemical transformations. The importance and problems of delivery and recovery systems will be emphasized. In-situ treatment technologies will be classified into extraction processes (air stripping, flushing, freezing, electrokinetics, vegetative and thermal) and immobilization and degradation processes (grouting, fixation, thermal fusion, chemical and biological). Documented case histories of an in-situ treatment at several sites will be presented. This will include some of the ongoing and completed EPA/ORD activities.

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Introduction

Management techniques for hazardous wastes found at Superfund sites may be grouped into two broad categories -- land disposal (either on-site or off-site) and use of technologies that permanently reduce or destroy the hazardous character of the waste. In general, land disposal has been used more frequently than other management techniques. This is because, for many waste types, land disposal is typically less expensive than technologies which change the nature of the waste. As a result, hazardous waste management at Superfund sites has frequently relied on relatively inexpensive land disposal. This reliance has limited the development and use of other options.

A review of 51 National Priority List (NPL) sites revealed that on-site land disposal, including capping and on-site landfilling, was used at 52 percent of the sites and off-site land disposal was used at 54 percent of the sites. A small percentage of the sites employed both techniques, by removing a portion of the contamination to off-site disposal while the remaining contamination was handled by on-site disposal.

Concern over the long-term reliability of land-based disposal practices has prompted elected and regulatory officials to develop more stringent controls on its use. The Resource Conservation and Recovery Act (RCRA) Amendments of 1984 included provisions to tighten the requirements governing land disposal of hazardous wastes. Under the Amendments, EPA is required to implement restrictions on the land disposal of hazardous wastes over a period of five to six years. Waste categories will be evaluated individually over this period to determine the exact nature of land disposal limitations for various waste types. EPA Superfund officials are also developing program policies to encourage the use of waste management techniques that are not dependent on land disposal practices.

In-situ treatment can be effective in eliminating or reducing the hazardous wastes at the site to acceptable levels. In-situ treatment of contaminated soils must be based on an understanding of factors and processes that determine the behavior of chemicals in soil systems. Specifically, an evaluation of chemical properties, biochemical processes, and environmental factors influencing the behavior and fate of chemicals is required. The goals of in-situ management include treating contaminated soils until acceptable levels of hazardous materials are achieved so that ground water and surface water resources are protected. In those cases where in-situ treatment is determined to be a feasible approach, it will usually be implemented as a part of an overall remedial action scenario involving the use of other methodologies (e.g. containment).

This paper will give a general overview/problem definition and state-of-the-art of the natural processes of biodegradation and chemical transformations. The importance and problems of delivery and recovery systems will be emphasized. In-situ treatment technologies will be classified into extraction processes (air stripping, flushing, freezing, electrokinetics, vegetative and thermal) and immobilization processes (grouting, fixation, thermal fusion, chemical and biological). Documented case histories of in-situ treatment at several sites will be presented. This will include some of the ongoing and completed EPA/ORD activities.

Much of this material has been derived from recent EPA documents (USEPA 1984a, USEPA 1985, USEPA 1986) and chapter four in the NATO-CCMS publication entitled "Contaminated Land Reclamation and Treatment" (Sanning 1985).

Definition of In-Situ Treatment

The term in-situ treatment of contaminated ground means that the treatment is applied without excavation of the material to be treated, irrespective of whether the waste is excavated as a total mass or removed in increments, and irrespective of whether the treated product is replaced or otherwise disposed of.

Conceptual possibilities include: grouting; liquid extraction using water or some other solvent; microbial degradation; chemical neutralization, fixation or degradation; and electro-osmosis. Most of these treatment systems have in common the need to inject fluids into the ground in one way or another, although less complex treatments such as deep ploughing to invert strata where contamination goes only to shallow depths is also a form of in-situ treatment. Some systems have the added potential advantage of improving some other property of the ground: for example, grouting may improve engineering properties.

Classifications of Treatment Methods

The potential methods of in-situ treatment can be classified in terms of their objectives:

1. To render the contamination harmless. This embraces such processes as microbial or chemical treatment to destroy

the contaminants and ground leaching to remove them physically.

2. 'Stabilization' in which the hazardous contaminants are rendered insoluble or otherwise immobile or their hazardous characteristics (e.g. toxicity or combustibility) are neutralized. The implication is that the converse may occur and that changing site conditions may in time 'destabilize' the contamination.

3. 'Solidification' involving the conversion of the contaminated ground into a solid mass - one beneficial effect of which may be stabilization of the contamination and a second an improvement in the engineering properties of the ground.

Classifications other than by objective as defined above are possible. For example:

1. in terms of the agent applied as follows;
   - chemical,
   - microbial,
   - solidification,
   - thermal.
2. in terms of method of application:
   - surface
   - ground leaching
   - ground injection
   - other (e. g. fusion/vitrification).

The following discussion addresses all three classifications.

Methods designed to isolate the contamination from any driving force that would cause the release or dispersal of hazardous components to the environment, i.e. 'macro-isolation' or 'macro-encapsulation' could be seen as a form of in-situ stabilization. This concept has not been used for the purposes of this study but it is important to recognize that the isolation of the site by the means of cut-off barriers, etc., may be an essential prerequisite of some in-situ treatments - for example, to confine a treatment agent within the site.

Treatment Technologies

In general the treatment technologies are adaptations or engineered processes to enhance natural methods for reduction of the concentration of hazardous compounds or reduction of the toxicity effect that the compounds may exhibit. Volatilization of organics or vacuum extraction of volatile compounds; soil washing with water, chelates, or surfactants may remove soluble inorganic or organic hazardous materials for subsequent treatment on or off-site; chemical neutralization, oxidation, or reduction can destroy specific hazardous substances, precipitation of metals with lime or sulfide can immobilize the metals and reduce toxicity; solidification either with cement grouts, asphalts, or by soil vitrification can also immobilize inorganic and organic compounds and reduce hazardous

effects; and biological forms such as plants, fungi, and bacteria can either accumulate certain metals or transform and destroy many hazardous organic compounds.

Delivery and Recovery Technologies

Delivery and recovery methods are critical for successful accomplishment of soil washing/flushing, chemical treatment, and biological treatment at the in-situ sites. An array of delivery techniques has been used for artificial ground water recharge and wastewater treatment by land application which may be applicable to waste deposit remediation efforts. These techniques can be utilized for applying or injecting water or chemical solutions into waste deposits in order to flush contaminants from waste deposits to the ground water table or react with contaminants in the waste deposit. Flushed contaminants can subsequently be collected and treated above ground. The available delivery methods are grouped into two generic categories: gravity and forced. In gravity applications, the flushing or reactant solution is applied directly over the waste deposit or delivered through the surrounding soil media to the waste deposit. After the solution percolates through the waste deposit, it enters the ground water. In forced methods, the flushing or reactant solution is directly injected into the waste deposit or surrounding soil medium through pipes by means of an applied pressure. Again, the solution, after passing through the waste deposit enters the ground water for subsequent recovery. A number of gravity delivery methods are available for selection including: flooding, ponding, surface spraying, ditch, infiltration galley and infiltration bed. The selection of a particular gravity delivery method would depend upon: surface topography, infiltration rate, configuration of waste deposit, ground water hydrology, hydraulic conductivity, soil porosity and local climate.

The forced delivery method would be cost-effective for a waste deposit and surrounding soil medium having a low permeability and low infiltration rate which precludes the delivery of treatment solution by natural gravity. Such systems would typically be comprised of an injection pipe (open end or slotted) and pump. The applicability of the forced delivery system would depend heavily on the site geohydrological conditions; thus, field boring tests and in-situ pumping tests should be conducted prior to the engineering and design of the forced delivery system.

Available recovery technologies can be categorized as gravity and forced. The recovery technologies discussed herein for in-situ treatment of waste deposits are those widely used in ground water recovery and construction dewatering operations.

Gravity recovery depends upon interception of the ground water, downgradient from the waste deposit. Thus, after applied treatment solutions pass through the waste deposit and enter the ground water, the resultant fluid is collected in an interceptor system (i.e., open ditch or buried drain) by simple gravity flow. Forced recovery systems utilize well points, deep wells or vacuum well points located in the ground water downgradient of the waste deposit to remove spent solutions.

The primary factors affecting the application of recovery methods are:

- Depth to ground water
- Geohydrologic properties of the waste deposit and surrounding soil media

Depth to ground water is a constraint for gravity systems only to the extent that there are practical limits to which excavation can be performed (typically less than 15 feet) (Husiman 1972) before costs become excessive or related widths of excavation become impractical. For forced systems, depth to ground water affects system installation costs and operational level energy consumption costs.

The primary geohydrologic property affecting recovery system application and feasibility is hydraulic conductivity. Hydraulic conductivity is related to the grain size distribution within the waste deposit and surrounding soil media. As hydraulic conductivity is reduced, the rate of spent solution recovery is decreased, thus increasing the period of spent solution recovery. In a related manner, forced recovery systems will require greater energy utilization as hydraulic conductivity decreases.

Special Method of Recovery Enhancement (Electro-Osmosis)

When a contaminated soil or waste exhibits a hydraulic conductivity of less than 10^{-5} cm/sec and the fluid to be extracted is high in inorganic constituents, some process must be used to assist the fluid in its travel through the waste deposit and surrounding media. Electro-osmosis is such a process.

Ground water migration by electro-osmosis is initiated by applying a direct-current electric potential to electrodes (anodes and cathodes) installed in the ground at a selected spacing within the low hydraulic conductivity media. The electric potential applied to the electrodes causes the positive ions in the pore water to move toward the cathodes. The movement of the positive ions develops tension in the media, causes the chemical composition of the media to change, forces the pore water to flow to the cathodes, controls the direction of the seepage forces and thereby reduces the water content of the deposit (Loughney 1975).

Some contaminated soil or wastes that are not able to be dewatered by wellpoints or wells, might be rendered drainable when these recovery methods are combined with a flow of direct electric current. By making the cathode a wellpoint, the water that migrates to the cathode can be removed by pumping. A site investigation should be performed prior to selecting any appropriate injection or recovery system. Once the site conditions are known and it has been determined that a special process is required to enhance hydraulic conductivity, conductivity tests should be performed to determine the electro-osmotic transmission coefficient, k_e (volume of water transmitted through a unit cross section in unit time by application of a potential of 1 volt/cm normal to the cross section).

Current requirements commonly range between 15 and 30 amp per

well, and power requirements are generally high. However, regardless of the expense of installation and operation of an electro-osmotic dewatering system, it may be the only effective means of dewatering and stabilizing certain fine-grained soils.

In an electro-osmotic dewatering system, the depth of the electrodes should be at least 5 ft below the bottom of the contaminated deposit that is to be dewatered. The spacing and arrangement of the electrodes may vary, depending on the configuration of the area to be dewatered and the voltage available at the site. Cathode spacings of 25 to 40 ft have been used, with the anodes installed midway between the cathodes. Electrical gradients of 1.5 to 4 volts per ft distance between electrodes have been successful in electro-osmotic stabilization. The electrical gradient should be less than about 15 volts per ft of distance between electrodes for long-term installations to prevent loss in efficiency due to heating the ground. Applied voltages of 30 to 100 volts are usually satisfactory; a low voltage is usually sufficient if the ground water has a high mineral content.

Electro-osmosis would only be technically feasible in waste sites having sufficiently low hydraulic conductivities. Hydraulic conductivities of less than 10^{-7} cm/sec prior to initiating electro-osmosis can be made to exhibit hydraulic conductivities in the range of 10^{-5} to 10^{-6} cm/sec for a gradient of one volt per ft using this method.

Use of long term, low power options should be considered when it is possible to do so within the time frame established for the site remediation.

Soil Flushing

Organic and inorganic contaminants can be washed from contaminated soils by means of an extraction process termed "soil flushing", "solvent flushing", "ground leaching", or "solution mining". Water or an aqueous solution is injected into the area of contamination, and the contaminated elutriate is pumped to the surface for removal, recirculation, or on-site treatment and reinjection. During elutriation, sorbed contaminants are mobilized into solution based on solubility, formation of an emulsion, or by chemical reaction with the flushing solution.

Solutions with the greatest potential for use in soil flushing fall into the following classes:

- Water
- Acids-bases
- Complexing and chelating agents
- Surfactants
- Certain reducing agents.

Water can be used to flush water-soluble or water-mobile organics

and inorganics. Hydrophilic organics are readily solubilized in water. Organics amenable to water flushing can be identified according to their soil/water partition coefficient, or estimated using the octanol/water coefficient. Octanol-water partition coefficients are available for a large number of compounds in: Substituent Constants for Correlation Analysis in Chemistry and Biology (Hansch and Leo, 1979). Chemical Property Estimation Methods (Lyman, Reehl and Rosenblatt, 1982) provides various methods for estimating the octanol-water partition coefficient using readily available physical and chemical data. Organics considered soluble in the environmental sense are ones with a partition coefficient (K) of approximately less than 1000 (log K = 3). High solubility organics, such as lower molecular weight alcohols, phenols, and carboxylic acids, and other organics with a coefficient less than 10 (log K $\leq$1) may have already been flushed from the site due to natural flushing processes. Medium solubility organics (K = 10 to 1000) which could be effectively removed from soils by water flushing include low to medium molecular weight ketones, aldehydes, and aromatics, and lower molecular weight halogenated hydrocarbons such as trichloroethylene (TCE) and tetrachloroethylene (PCE). Inorganics which can be flushed from soil with water are soluble salts such as the carbonates of nickel, zinc, and copper. Adjusting the pH with dilute solutions of acids or bases (to be discussed) will enhance inorganic solubilization and removal.

The remedial actions at the Goose Farm site in Plumstead, NJ, involved the installation of a wellpoint collection and recharge system and the flushing of contaminants from the soil with water (USEPA, 1984b). Water flushing has been used over a 5 to 6 year period to reclaim a former herbicide factory site in Sweden (Truett, Holberger, and Sanning, 1982). Cleanup of a TCE spill in Germany involved water flushing of contaminated soils, and effected a 50 percent decrease in TCE over an 18 month period (Stief, 1984).

Dilute solutions of acids have been widely used in industrial processes to extract metal ions. Solutions of sulfuric, hydrochloric, nitric, phosphoric, and carbonic acid are used in industrial applications to dissolve basic metal salts (hydroxides, oxides, and carbonates). However, because of the toxicity of many acids, it is desirable to use weak acids for in-situ treatment. Sodium dihydrogen phosphate (NaH_2PO_4) and acetic acid (CH_3COOH) have low toxicity and are reasonably stable. A stronger dilute acid such as sulfuric acid would be used if the soil contained sufficient alkalinity to neutralize it. Acidic solutions may also serve to flush some basic organics such as amines, ethers, and anilines.

Complexing and chelating agents may also find use in a solution mining removal system for heavy metals. Some commonly employed substances are citric acid, ethylenediaminetetracetic acid (EDTA), and diethylenetriaminepentacetic acid (DTPA) (Rogoshewski and Carstea, 1980). Chelating agents used for in-situ treatment must result in a stable metal-chelate complex which is resistant to decomposition and degradation.

Another possibility for mobilizing metals which are strongly adsorbed to manganese and iron oxides in soils is to reduce the metal

oxides which results in release of the heavy metal into solution. Chelating agents or acids can then be used to keep the metals in solution. Treatment agents which may be suitable for this purpose include hydroxylamine together with an acid, or sodium dithionite/citrate.

Surfactants can be used to improve the solvent property of the recharge water, emulsify nonsoluble organics, and enhance the removal of hydrophobic organics sorbed onto soil particles. Surfactants improve the effectiveness of contaminant removal by improving both the detergency of aqueous solutions and the efficiency by which organics may be transported by aqueous solutions. A major problem with surfactants is that once they have been used to solubilize the contaminants, the recovered solutions are very difficult to treat.

Numerous environmentally safe and relatively inexpensive surfactants are commercially available. Use of surfactants to date has been restricted to laboratory research. Most of the research has been performed by the petroleum industry for tertiary oil recovery (Barakat et al., 1983; Cash et al., 1977; Doe, Wade, and Schechter, 1977; and Wilson and Brandner, 1977). Aqueous surfactants have also been proposed for gasoline cleanup. In a study performed by the Texas Research Institute (1979) for the American Petroleum Institute (API), a mixture of anionic and nonionic surfactants allowed contaminant recovery of up to 40 percent. In a laboratory study conducted by Ellis and Payne (1983), crude oil recovery was increased from less than 1 percent to 86 percent, and PCB recovery was increased from less than 1 percent to 68 percent when soil columns were flushed with an aqueous surfactant solution. Even if the process of surfactant washing could be optimized to removal efficiencies of the order of 90% or better, it would probably not be able to meet applicable and relevant standards for cleanup. This stresses the fact that no single unit operation will achieve the necessary final cleanup levels. Only a combination of well designed and engineered unit operations which go together to constitute the "remedial action scenario" can be expected to succeed.

The advantages of the soil flushing processes are that, if the waste is amenable to this technique and distribution, collection, and treatment costs are relatively low, solution mining can present an economical alternative to the excavation and treatment of the wastes.

Microbial Treatment

Bioreclamation is a technique for treating zones of contamination by microbial degradation. The basic concept involves altering environmental conditions to enhance microbial catabolism or cometabolism of organic contaminants, resulting in the breakdown and detoxification of those contaminants. The technology has been developed rapidly over recent years, and bioreclamation appears to be one of the most promising of the in-situ treatment techniques.

Considerable research conducted over the past several decades has confirmed that microorganisms are capable of breaking down many of those organic compounds considered to be environmental and health hazards at spill sites and uncontrolled hazardous waste sites; laboratory,

pilot, and field studies have demonstrated that it is feasible to use this capability of microorganisms in-situ to reclaim contaminated soils and ground water.

The bioreclamation method that has reached the most advanced stage of development for in-situ treatment is one which relies on aerobic (oxygen requiring) microbial processes. Enhanced indigenous microbial degradation involves optimizing environmental conditions by providing an oxygen source and nutrients which are delivered to the subsurface through an injection well or infiltration system to enhance microbial activity. Indigenous microorganisms can generally be relied upon to degrade a wide range of compounds given proper nutrients and sufficient oxygen. Specially adapted or genetically manipulated microorganisms are also available and may be added to the treatment zone (see Page 10).

For most compounds, the most rapid and complete degradation occurs aerobically. There are some compounds, most notably the lower molecular weight halogenated hydrocarbons, which will only degrade anaerobically. [However, recent research conducted at the USEPA Robert S. Kerr Laboratory has discovered degradation of TCE in the presence of oxygen and methane gas (Wilson 1985).]

Anaerobic microorganisms are also capable of degrading certain organic contaminants. Methanogenic consortiums, groups of anaerobes that function under very reducing conditions, are able to degrade halogenated aliphatics (e.g., TCE) while aerobic organisms cannot. The potential for anaerobic degradation has been demonstrated in numerous laboratory studies and in industrial waste treatment processes that use anaerobic digesters or anaerobic waste lagoons as part of the treatment process. Using anaerobic degradation as an in-situ reclamation approach is theoretically feasible.

The feasibility of bioreclamation as an in-situ treatment technique is dictated by waste and site characteristics. More specifically, those factors which determine the applicability of a bioreclamation approach are:

- Biodegradability of the organic contaminants
- Environmental factors which affect microbial activity
- Site hydrogeology.

Bioreclamation can be expected to reduce the concentration of only those organic compounds which are amenable to biological degradation. These are compounds that are either substrates for microbial growth and metabolism (the organism uses the compound as a carbon and energy source), or are cometabolically broken down as the microorganism uses another primary substrate as its carbon and energy source.

An extensive review of research on the relative biodegradabilites of environmental pollutants has been gathered in a document (USEPA 1985). Relative aerobic biodegradability can also be estimated using laboratory data associated with oxygen requirements for decomposition

(i.e. 5-day biological oxygen demand [BOD_5] or a measurement of continuous oxygen uptake rate with the Sapromat equipment).

One of the best documented case histories of biodegradation treatment is the case at Biocraft Laboratories in Waldwick, New Jersey (USEPA 1984b).

Biocraft Laboratories is a small synthetic penicillin manufacturing plant located on a 4.3 acre site in an industrial park in Waldwick, NJ. In August 1975, contamination was observed in a small creek which was traced to a storm sewer draining the site and subsequent investigations revealed a leak in underground storage process lines connecting solvent storage facilities. The inventory of contaminants estimated to have escaped into the environment is presented as follows:

TABLE 1. BIOCRAFT SITE ESTIMATED CONTAMINATION INVENTORY IN SOILS

Methylene Chloride	CH_2Cl_2	181500 lb
N-Butyl Alcohol	$C_4H_{10}O$	66825 lb
Dimethyl Aniline	$C_8H_{11}N$	26300 lb
Acetone	C_3H_6O	10890 lb

Driven by the high cost and ongoing uncertainty associated with the effectiveness of the initial remedial method in treating the contaminant, Biocraft developed as alternative: collecting the contaminant plume down gradient in a slotted pipe collection trench, treating the collected ground water in a surface aerobic biological treatment system, injecting the treated "bioactive" water upgradient in two slotted pipe recharge trenches and enhancing in-situ biological treatment in the subsurface through injection of air in equally spaced air injection wells.

A summary of the performance is given as follows:

AVERAGE REMOVAL EFFICIENCY OF AEROBIC TREATMENT SYSTEM (3)

Target Contaminant	Reported Removal Efficiency
Methylene Chloride	98%
n-Butyl Alcohol	98%
Acetone	97%
Dimethyl Aniline	93%

It is reported that the overall remediation at the site is proceeding successfully.

Vegetational Uptake

Plant uptake of an element is a function of four processes and their interrelationships. These processes include availability of the element in the soil, movement of the element to the root, absorption of the element by the root (roots absorb only ionic forms of elements) and translocation of the element in the plant.

There is some evidence from research studies that certain contaminants in soil will be taken up and translocated into growing plants. Repeated growth, harvest, and regulated disposal of the plants may have the potential for removing contaminants from the soil. Removal is highly variable and depends on the soil type, the specific contaminant and concentration level, and the crop used. In various parts of the world plants can be found that have developed the ability to accumulate specific elements to an extent that would generally be lethal to most plants. The ash of the plant may, in rare instances, contain percentage amounts of the elements, but in most cases the degree of concentration is much less.

The first steps in the development of a vegetative uptake system are as follows:

1. Determine whether vegetative extraction from the contaminated soil has a high probability of being the most technically and cost effective approach at a specific site, realizing that this approach will require a substantial time period and intensive agronomic management over that time.
2. Determine whether suitable plant species (or varieties within species) are available to accomplish the desired contaminant extraction.
3. Determine whether the site possesses, or can be readily modified to possess, soil conditions which will support optimal growth of the selected plant materials.
4. Conduct greenhouse-scale confirmatory uptake tests.
5. Confirm that the plant materials which have extracted soil contaminants can be adequately disposed of in an environmentally safe manner, and that the plant mass and harvesting mechanics are realistically manageable.

Although there are plant species that are capable of measurable extraction of numerous inorganic soil contaminants, such as heavy metals and elements metabolized similarly to nutritional elements, there is little evidence as yet regarding the potential of this approach in soils containing contaminants such as hydrocarbon solvents and refractory organic compounds. A few investigators have proposed studies parallel to those which have confirmed the uptake and metabolism of soil-applied herbicides such as those commonly used in agricultural practice. However, since many organic pollutant molecules are substantially different in structure and reactivity from agricultural herbicides, further research is essential to confirm the utility of this concept. Dr. Robert Bell of the University of Liverpool is currently conducting a study on the uptake of organic pollutants by plants at the USEPA Test and Evaluation (T & E) facility in Cincinnati, Ohio.

Chemical Treatment

In-situ treatment techniques discussed in this section are those which serve to destroy, degrade, or otherwise reduce the toxicity of contaminants and include neutralization, hydrolysis, oxidation/reduction, enzymatic degradation, and permeable treatment beds. These methods

are applicable to specific chemical contaminants; therefore, use of these in-situ techniques at waste sites will be limited.

Neutralization involves injecting dilute acids or bases into the ground water to adjust the pH. This pH adjustment can serve as pretreatment prior to in-situ biodegradation, oxidation, or reduction to optimize the pH range. It can be used to neutralize acidic or basic plumes that need no other treatment, or to neutralize ground water following another treatment. It can also be used during oxidation, reduction, or precipitation to prevent the formation of toxic gases including hydrogen sulfide and hydrogen cyanide.

The pH adjustment can also be used to increase the hydrolysis rate of certain organics. Of the parameters which affect the rate of hydrolysis (temperature, solvent composition, catalysis, and pH), pH adjustment has the greatest potential. The rate of hydrolysis can be increased up to one order of magnitude for a change of one standard unit in pH (USEPA, 1985). Classes of compounds with potential for in-situ degradation by hydrolysis include:

- Esters
- Amides
- Carbamates
- Phosphoric and phosphonic acid esters
- Pesticides.

Because a hydrolysis product may be more toxic than the present compound, the pathways for reactions must be determined to ensure toxic products are not produced. USEPA (1985) has a more thorough discussion of this technology.

Oxidation and reduction reactions serve to alter the oxidation state of a compound through loss or gain of electrons, respectively. Such reactions can detoxify, precipitate, or solubilize metals, and decompose, detoxify, or solubilize organics. Oxidation may render organics more amenable to biological degradation. As with many of these chemical treatment technologies, oxidation/reduction techniques are standard wastewater treatment approaches, but their application as in-situ treatment technologies is largely conceptual.

Oxidation of inorganics in soils is for all practical purposes limited to oxidation of arsenic and possibly some lead compounds. The in-situ oxidation of arsenic compounds with potassium permanganate ($KMnO_4$) has been used to successfully reduce the arsenic concentrations in ground water in the vicinity of a zinc ore smelter near Cologne, Germany (Stief, 1984). 64,000 lb. of $KMnO_4$ were injected into 17 wells and piezometer wells resulting in an average decrease in arsenic ground water concentrations from 13.6 mg/l to 0.06 mg/l from 1975 to 1977. In 1979, however, an increase indicated that the mixing of ground water and $KMnO_4$ had not been thorough.

Of the numerous oxidizing agents available, three have been considered potentially useful in the in-situ detoxification of organics, ground water and soils contaminated with organics: hydrogen peroxide, ozone, and hypochlorites (USEPA, 1985). Each can react with a broad range of organics and could potentially oxidize a number of different organic contaminants in a hazardous waste site. Selection of the appropriate oxidizing agent is dependent in part upon the substance or substances to be detoxified, but also upon the feasibility of delivery and environmental safety. There are some compounds that will not react with hydrogen peroxide but will react with ozone or hypochlorite.

Ozone gas is a very strong oxidizing agent that is very unstable and extremely reactive. It cannot be shipped or stored; therefore it must be generated on-site prior to application. Ozone rapidly decomposes and its half-life in ground water is only 18 minutes (USEPA, 1985). Ozone is used in the treatment of drinking water, municipal wastewater, and industrial waste, and has been used in the treatment of contaminated soils and ground water at the Karlsruhe site in Germany (Rice 1984). A well field which served as a drinking water supply for over 500,000 people was rejuvenated. Ozone oxidation with ultraviolet irradiation has been used to successfully reduce concentrations of benzene, phenols, and trichloroethylene in lake water.

Hypochlorite, generally available as potassium, calcium, or sodium hypochlorite (bleach) is also used in the treatment of drinking water, municipal wastewater, and industrial waste. Hypochlorites have never been used in the treatment of contaminated ground water or soils. Tolman et al., (1978) has described the conceptual design and in-situ detoxification of cyanide with sodium hypochlorite. The reaction of many organics with hypochlorite results in the formation of chlorinated organics which can be as or more toxic than the original contaminant. The formation of lower molecular weight chlorinated organics, (e.g., trihalomethanes) in drinking water from hypochlorite treatment for disinfection purposes has become a major concern of the drinking water industry.

Hydrogen peroxide (H_2O_2), a moderate strength chemical oxidant, is used routinely in municipal wastewater treatment to control various factors of biological treatment, and is also used in industrial waste treatment to detoxify cyanide and various organic pollutants. It is commercially available in aqueous solutions of several concentrations and is miscible in water at all concentrations. It has been delivered successfully in dilute solutions to the subsurface as an oxygen source in a bioreclamation project (Raymond, Jamison, and Hudson, 1984). Several suppliers have developed a line of hydrogen peroxide solutions specifically designed for reclamation purposes.

Chemical reduction is the process by which the oxidation state of a compound is reduced. Reducing agents are electron donors, with reduction accomplished by the addition of electrons to the atom.

Chemical reduction does not appear as promising as oxidation for the treatment of organics. Although researchers have demonstrated reductive dehalogenation of a variety of chlorinated organics and

reduction of unsaturated aromatics and aliphatics in laboratory studies using catalyzed metal powders, the treatment reagents are costly and the effectiveness of chemical reduction in soils remains to be demonstrated.

Chemical reduction does, however, appear promising for treatment of chromium and selenium in soils. The in-situ reduction of hexavalent to divalent chromium has been accomplished in Arizona well water using minute quantities of reducing agent. (Srivastava and Haji-Djafari, 1983).

There are a number of disadvantages with the use of oxidizing and reducing agents which limit their use at hazardous waste sites. The treatment compounds are non-specific and this may result in degradation of non-targeted compounds. There is the potential, particularly with oxidation, for the formation of more toxic or more mobile degradation products. Also, the introduction of these chemicals into the ground water system may create a pollution problem in itself. As with soil flushing, uncertainty exists with respect to obtaining adequate contact with the contaminants in the plume.

Enzymatic degradation of organics with cell-free enzymes holds potential as a possible in-situ treatment technique. Purified enzyme extracts, harvested from microbial cells, are commonly used in industry to catalyze a variety of reactions, including the degradation of carbohydrates and proteins. A bacterial enzyme preparation has been used to detoxify organophosphate waste from containers (Munnecke, 1980). Parathion hydrolase has been tested under field conditions in the degradation of the pesticide diazinon (Paulson et al., 1984). The studies indicate that parathion hydrolase can be used to effectively reduce rapidly large concentrations of diazinon in soil. The enzyme is readily soluble in water, is reasonably stable at summer temperatures, and can be easily handled in the field. The pH or organic content of the soil does not appear to affect the enzyme's effectiveness. It appears from the study that parathion hydrolase could be used effectively to clean up diazinon spills, but more research is required to determine how to obtain optimal field efficacy of the enzyme.

Immobilization

Immobilization methods are designed to render contaminants insoluble and prevent leaching of the contaminants from the soil matrix and their movement from the area of contamination. Little is currently known about the effectiveness and reliability of immobilization techniques (Truett, Holberger, and Sanning, 1982).

Immobilization methods which are currently being investigated include: precipitation, chelation, and polymerization.

Precipitation is the most promising method for immobilizing dissolved metals such as lead, cadmium, zinc, and iron. Some forms of arsenic, chromium, mercury, and some organic fatty acids can also be treated by precipitation (Huibregtse and Kastman, 1979). All of the divalent metal cations can be precipitated using sulfide, phosphate, hydroxide, or carbonate. However, the solubility product and the stability of the metal complexes vary. Because of the low solubility

product of sulfides and the stability of the metal sulfide over a broad pH range, sulfide precipitation looks most promising. The remaining anions decrease in effectiveness in the following order: phosphate > hydroxide > carbonate. Metal carbonates and hydroxides are stable only over a narrow pH range and the optimum pH range varies for different metals. Precipitation of the metal as the metal phosphate may require very high concentration of orthophosphate since calcium and other naturally occurring soil cations present in high concentrations will precipitate first.

Sodium sulfide used in conjunction with sodium hydroxide has shown widespread applicability for precipitation of metals. Precipitation takes place at a neutral or slightly alkaline pH. Resolubilization of sulfides is low. Addition of sodium hydroxide minimizes the formation of hydrogen sulfide gas by assuring an alkaline pH. Experiments with sulfide precipitation of zinc indicate that a high residual of unreacted sulfide may remain in solution.

As with other in-situ techniques, precipitation is most applicable to sites with sand or coarse silt strata. Disadvantages include the injection of a potential ground water pollutant; the potential for formation of toxic gases (in the case of sulfide treatment); the potential for clogging soil pore space; and the possibility of precipitate resolubilization.

The use of chelating agents may also be a very effective means of immobilizing metals although considerable research is needed in this area. Depending upon the specific chelating agent, stable metal chelates may be highly mobile or may be strongly sorbed to the soil. Tetran is an example of a chelating agent which is strongly sorbed to clay in soils (USEPA, 1984a).

A third method for immobilizing metals applies specifically to chromium and selenium. These metals can be present in the highly mobile, hexavalent state but can be reduced to less mobile Cr (III) and Se (IV) by addition of ferrous sulfate. Arsenic exists in soils as either arsenate, As (V), or as arsenite, As (III), the more toxic and soluble form. Arsenic can be effectively immobilized by oxidizing As (III) to As (IV) and treating the As (IV) with ferrous sulfate to form highly insoluble Fe_2AsO_4.

Polymerization involves injection of a catalyst into a ground water plume to cause polymerization of an organic monomer (e.g., styrene, vinyl chloride, isoprene, methyl methacrylate, and acrylonitrile). The polymerization reaction transforms the once fluid substance into a gel-like, non-mobile mass. In-situ polymerization is a technique most suited for ground water cleanup following land spills or underground leaks of pure monomer. Applications for uncontrolled hazardous waste sites are very limited. Major disadvantages include very limited application and difficulty of initiating sufficient contact of the catalyst with the dispersed monomer (Huibregtse and Kastman, 1979). In-situ polymerization was successfully performed to remedy an acrylate monomer leak, in which 4,200 gallons of acrylate monomer leaked from a corroded underground pipeline into a glacial sand and gravel layer.

Soil borings indicated that as much as 90 percent of the monomer had been polymerized by injection of a catalyst, activator, and wetting agent (Williams, 1982).

Stabilization/Solidification

These terms describe a process whereby the physical characteristics of a waste can be altered to reduce the mobility of its hazardous constituents as well as improve waste handling. Stabilization/solidification entails mixing a hardening agent, called a fixative, with the waste. There are three possible mechanisms by which the contaminants can be held: weak Vander Waals forces, strong chemical bonds between the fixative and waste contaminants, and microencapsulation whereby the contaminants are individually encased in the fixative. It is believed that most fixatives in current use stabilize contaminants through microencapsulation.

Nevertheless, the end-product should possess the following properties:

1. Reduced water permeability through the waste:
2. Decreased surface area for waste contact with leaching medium;
3. Reduced solubility of hazardous constituents to leaching medium;
4. Increased permanent binding structure of waste material (better waste handling); and
5. Decreased toxicity of hazardous constituents.

Finally, the decision to use a stabilization technique should be made only after the chemical and physical properties of the solidified waste have been extensively tested to insure that the required properties have been developed. Currently, there is no standard testing program other than waste delisting for classifying or determining the suitability of a solidified waste for disposal. Consequently the data that exists on stabilized products has limited value because of the incompleteness of data and the variability in testing techniques. It is also difficult to generalize because each testing program must be tailored to take into account the type of fixative, and type of waste. Common physical parameters measured include bulk and dry unit weight, unconfined general compressive strength, permeability, wet/dry durability, and freeze/thaw durability. Note that organic fixatives are not tested for unconfined compressive strength because they usually require secondary containment. The chemical stability of a waste is determined by leach testing of the waste, i.e. a variation of the EP toxicity test.

Grouting in Contaminated Soils

The injection of gelling and solidifying agents for engineering purposes is known as 'grouting'. Grouting is frequently used to reduce the permeability of a mass of material or to form cut-off barriers to eliminate water flow beneath dams or adjacent to engineering structures.

The technical difficulties of grouting to contaminated sites, are indicative of the problems of ground injection techniques in general as most employ similar equipment and must cope with similar physical difficulties.

When grouting is applied to treat contaminated soils, it is often necessary to inject at shallow depths (less than 2 m), since the contaminated volume is generally in the soil mass immediately below the surface. One of the basic characteristics of grouting is the need for adequate overburden pressure to ensure that the grout does not escape to the surface through fissures before it permeates the target areas. To overcome this problem, two solutions have been suggested:

1. Applying a temporary surcharge, such as a suitable concrete block (placed on the surface and moved about, as necessary, by crane) or a mound of clean fill material.

2. The use of electro-osmosis to improve the receptivity of the soil to grouting, or as part of the grouting process. It has been shown that the presence of an electric field reduces the grouting pressures necessary and induces movement of the grout. The electric energy requirements are not well established but appear modest - in the range of 2 to 8 kWh per cubic meter of material.

Stabilization by the Action of Ion Exchange Resins

Wastes containing heavy metals and certain organic compounds can produce leachate contaminated by toxic cations (e.g. heavy metals) and anions (e.g. cyanide). When such leachates contact selected ion exchange resins under appropriate conditions, the toxic ions in the leachate will attach to the resin while releasing an innocuous ion into the leachate. A related approach that might prove useful for contaminated land is to inject into the contaminated ground, a fluid containing ion exchange materials and a material that will polymerize or form a gel. Under appropriate conditions, the polymer or gel will form in-situ and will be embedded with the ion exchange resin. The solid component (e. g. the polymer or gel, or a matrix of these materials and soil) will tend to impede movement of leachate through the soil while immobilizing toxic species in the leachate through the mechanism of ion exchange. Laboratory experimentation with this process indicated it to be technically feasible, but field demonstrations have not yet been reported (Porter 1981).

Information on contamination is needed to determine whether there will be any interference with the proper 'setting up' of a solidification material by crystallization, polymerization, gelling, adhesion, or other setting mechanisms. It is also needed to permit formulation of the proper additives for neutralization or detoxification, if this approach is used. The required level of detail concerning contamination and physical characteristics differs for the various processes.

In the discussion that follows, it is assumed that the required information about the contaminated land, surrounding rock and soil, and

ground water, is available or can be determined at the required level of detail by established methods of sampling and analysis. This information may include chemical composition and physical characteristics of the waste or soil; quantities and spatial distribution of the contaminants; types and characteristics of surrounding and intermixed soils, rocks, and other geological materials; and rates and directions of ground water movement. Methods for acquiring these types of information are described in a variety of manuals, handbooks, and other sources (EPA 1979, EPA 1980).

The second requirement, that of providing a sufficient degree of mixing or contact between the contaminants and the treatment agent(s), presents a more fundamental and difficult problem in the general case of multiple-constituent wastes buried and intermixed with soils of different textures and hydraulic conductivities. A considerable amount of information on necessary contact time between reactive agent(s) and material to be treated has been established in trials of various solidification and stabilization processes for hazardous wastes. For example, in some processes such as containment of the solid within a bituminous material, the efficiency of the process (in terms of amount of waste contained per unit of solid formed) depends on the thoroughness of the chemical reactions that form the solid product (as in providing water of hydration in a cement setting process), or when two or more additives must be blended (as with a polymerizing agent and a monomer in addition to the waste), the thoroughness of the mixing and blending is critical to the formation of a solid with the desired properties.

The degree of mixing or contact that can be expected to occur when a treatment agent is applied in-situ to a given landfilled waste or contaminated soil is dependent on a large number of parameters including:

- viscosity of the applied agent
- permeability of the waste material
- permeability of surrounding soils (if they are contaminated)
- porosity of the waste material and soils
- degree of saturation of waste material and soils
- spatial distribution of the waste material relative to soils, rocks, and other surrounding materials
- setting time for solidification agents.

Surface Application

The stabilizing agent can be applied to the land surface in solid or liquid form and allowed to penetrate the waste materials or contaminated ground by infiltration, provided the ground is sufficiently permeable (and provided a chemical applied to the surface in solid form is dissolved or otherwise transported by rainfall or wetting). Surface application would be appropriate primarily for very shallow, permeable, loosely-packed fills in which simple chemical reactions can be used for rendering a specific waste component less hazardous. It does not appear generally useful where a buried waste is to be incorporated or enclosed in a rigid solid mass.

Vitrification

A thermal treatment process is currently under study to provide additional environmental protection of transuranic and chemically contaminated soil sites. The process is known as in-situ vitrification (ISV). In-situ vitrification is a remedial action process that destroys solid and liquid organic contaminants and incorporates radionuclides into a glass-like material that renders contaminants substantially less mobile and less likely to impact the environment. A large-scale operational acceptance test (LSOAT) was recently completed in which more than 180 t of vitrified soil were produced in each of three adjacent settings (Buelt 1986). The method has been developed by Battelle Memorial Institute Pacific Northwest Laboratory in the state of Washington for treatment of soils contaminated with radioactive wastes. However, the method may be applicable to soils containing a variety of hazardous wastes.

A number of methods are currently being developed which involve physical manipulation of the subsurface in order to immobilize or detoxify waste constituents. These technologies, which include in-situ heating, and ground-freezing, are in the early stages of development.

In-situ heating has been proposed as a method to destroy or remove organic contaminants in the subsurface through thermal decomposition, vaporization, and distillation. Methods recommended for in-situ heating are steam injection (Hoogendorn, 1984) and radio frequency heating (Dev, Bridges, and Sresty, 1984).

Radio Frequency Heating

The radio frequency heating process has been under development since the 1970s. Field experiments have been conducted for the recovery of hydrocarbons. The method involves laying a row of horizontal conductors on the surface of a landfill and exciting them with an RF generator through a matching network. The decontamination is accomplished in a temperature range of 300° to 400°C, assisted with steam, and requires a residence time of about two weeks. A gas or vapor recovery system is required on the surface. Excavation, mining, drilling, or boring is not required. Preliminary design and cost estimates for a mobile RF in-situ decontamination process indicate that the method is 2 to 4 times cheaper than excavation and incineration (Dev, Bridges, and Sresty, 1984). This method appears very promising for certain situations involving contamination with organics, although more research is necessary to verify the effectiveness in-situ.

Ground Freezing

Artificial ground freezing involves the installation of freezing loops in the ground and a self-contained refrigeration system that pumps coolant around the freezing loop (Sullivan, Lynch and Iskandar, 1984). Although never used in an actual waste containment operation, the technology has been used extensively as a construction method in civil engineering projects. Artificial ground freezing has not yet been aplied to the waste itself, which may have a freezing point much

lower than that of the soil systems. Freezing the soil surrounding the hazardous waste would render the soil practically impermeable. It may be useful only as a temporary treatment approach because of the thermal maintenance expense (Sullivan, Lynch, and Iskandar, 1984).

Air Stripping

Contaminated ground waters and landfill leachates containing volatile organic compounds have been successfully remediated by air stripping at a number of industrial and Superfund sites. In addition, mechanical aeration systems are being developed for removal of volatiles from soils at selected sites.

In-situ air stripping involves the removal of volatile organics from a soil matrix by mechanically drawing or venting air through the unsaturated soil layer. The soils are gradually treated as the volatile organics are partioned from the soil particles, organic matter and soil moisture are driven into the air phase within the soil pore spaces. The in-situ air stripping system must be designated and operated in accordance with site specific subsurface conditions to maximize the contaminant removal effectiveness (Corbin, Anastos, Coia and Koltuniak 1986).

Design and operation of an in-situ stripping system is based upon the theory of gas movement in porous media. Air stripping applications at hazardous waste sites must address the chemical characteristics of contaminant migration as well as the characteristics of air flow and pressure in the subsurface soils. In-situ air stripping has received limited attention as a potential remedial action alternative, and no design or operating parameters have been established in previous investigations. The basic theory of subsurface gas movement, such as the design theory for landfill methane recovery, was utilized as the framework for pilot systems. TCE has been effectively removed from the soils through the use of forced air ventilation systems. Some potential limitations should be noted, since additional investigation is required in the following general areas:

- The effects of various soil types and the presence of perched water zones on air stripping effectiveness is unknown.
- Detailed laboratory-scale investigations may be used to evaluate contaminant migration potentials for varying soil types and chemical constituents.

SITE Program

EPA's Office of Emergency and Remedial Response (OERR) has recently issued guidance to increase the use of alternative technologies at Superfund sites, and in conjunction with the Office of Research and Devlopment (ORD), has established a formal program to enhance the development and use of new or innovative technologies for this purpose. The program is called the Superfund Innovative Technology Evaluation or SITE Program.

The primary purpose of the Program is to enhance the development

and demonstration, and thereby establish the commercial availability, of innovative technologies at Superfund sites as alternatives to the containment systems presently in use. The program is a joint effort between ORD and the OERR.

There will be four parts to the SITE Program:

1. Identify, and remove where possible, impediments to the development and commercial use of alternative technologies.

2. Conduct a demonstration program of the more promising innovative technologies to establish reliable performance and cost information for site cleanup decision making.

3. Develop procedures and policies which encourage selection of available alternative treatment remedies at Superfund sites.

4. Structure a research and development program that will nurture emerging technologies.

EPA has recognized that there are a number of forces which are inhibiting the expanded use of alternative technologies at Superfund sites. The first part of the Program will include the identification of both incentives and disincentives that may be implemented or removed as appropriate. Three types of problems have been identified: informational, regulatory, and institutional.

Shortages of information regarding testing and evaluation methodologies, costs, and available markets have been recognized as informational impediments. Regulatory incentives and disincentives may result from EPA policies, regulations and procedures as well as non-EPA legislative and regulatory requirements. Evaluation of institutional problems will include such items as public acceptance of innovative technologies, economic factors, and liabilities assumed by developers and users of a new technology. In each case, barriers to be overcome and methods to promote the use of alternative technologies will be evaluated as appropriate. It should be noted that EPA intends to phase this study so that incentives and impediments which are under EPA's control may be identified first. Those that will require action outside of EPA will be addressed later. In this way, EPA will be able to act on those areas in which it has influence early in the overall program.

The second part of the SITE Program will involve the demonstration and evaluation of selected technologies. This is intended to be a significant ongoing effort requiring the participation of ORD, OERR, and the private sector. The objective of the demonstration program is to evaluate fully developed technologies so that cost effectiveness information will be available. Superfund decision-makers will thus have the necessary information to effectively consider these technologies for future cleanup projects. Consistent with pending legislation, it is planned that the first round of demonstrations will include at least 10 technologies.

The demonstration program will involve periodic solicitation and selection of candidate alternative technologies for Superfund sites. During this phase, ORD will solicit proposals to demonstrate existing and developed technologies and will screen those proposals for inclusion in the demonstration program. In addition, OERR will screen various Superfund wastes and sites to select those that represent a high priority for demonstration. The two efforts will be combined to result in the final selection of approximately 10 demonstration projects that will represent various combinations of priority waste sites and innovative alternative technologies. Additional activities will include development of testing, QA/QC, and data evaluation procedures. ORD and OSWER will then work with the inventor or developer, as appropriate, to conduct a demonstration. It is EPA's intent to have the private sector conduct the demonstration, with EPA evaluating the results. The end result will be (1) a series of reports issued by EPA evaluating specific technologies and their applicability to Superfund site cleanups and (2) a companion technology transfer program.

During the initial selection phase, EPA intends to solicit public comment and conduct an outreach program to keep all interested parties informed of progress in the demonstration program. In addition, once a specific demonstration is being considered, EPA will establish a community relations program to allow for input from the public in the area surrounding the demonstration site. After a demonstration is completed, EPA will ensure that the results are made available to all interested parties.

As information becomes available from the demonstration program, EPA will evaluate those results in the context of existing policies and procedures to determine how the successful technologies can best be used in the Superfund program.

The final part of the SITE Program will encourage further development of promising innovative technologies that are not presently ready for demonstration. This effort will be conducted by ORD. Activities will involve ensuring that EPA is aware of advances in the state-of-the-art for alternative technologies and, using existing mechanisms, selecting emerging technologies for EPA support to accelerate and promote their development.

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